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Evolution of dislocation structure in neutron irradiated Zircaloy-2 studied by synchrotron x-ray diffraction peak profile analysis

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ABSTRACT

Dislocation structures in neutron irradiated Zircaloy-2 fuel cladding and channel material have been characterized by means of high-resolution synchrotron x-ray diffraction combined with whole peak profile analysis and by transmission electron microscopy (TEM). The samples available for this characterization were taken from high burnup fuel assemblies and offer insight into the evolution of the dislocation structure after the formation of dislocation loops containing a *c* component. Absolute dislocation density values are about 4–15 times higher for the whole peak profile compared to TEM analysis. Most interestingly, the diffraction analysis suggests that the total dislocation density, as well as the *a* loop density, increases with fluence for the cladding material type. This trend is also inferred from a Williamson-Hall representation but contradicts the TEM observations. The *c* loop density evolution is more complicated and doesn't display any particular trend. In addition, the diffraction analysis highlights the presence of well-developed shoulders adjacent to the basal reflections and noticeable peak asymmetry particularly for the channel samples that experienced slightly lower operation temperatures than the clad. The findings are discussed in respect of the perceived irradiation induced growth mechanisms in Zr alloys.

1. Introduction

Zirconium alloys are widely used by the nuclear industry as fuel cladding and reactor core structural material due to their low neutron absorption cross-section, suitable mechanical properties and good corrosion resistance. Despite these advantageous characteristics, zirconium cladding exhibits dimensional instabilities in the high temperature irradiation environment of a nuclear reactor, which are a result of irradiation induced growth, irradiation enhanced creep and hydrogen pick up [1–3]. Irradiation growth is, in the case of zirconium fuel cladding with a typical split basal texture, the volume conserving axial expansion and radial contraction of the clad under fast neutron irradiation ($E > 1$ MeV) that is independent of applied stress. Irradiation growth has been observed to be dependent on neutron fluence [4], temperature [5,6], alloy composition [7] and thermomechanical history (crystallographic texture and degree of dislocation density in the processed material) [8,9] but a detailed mechanistic understanding is presently absent.

In recrystallised zirconium alloys with a typical split basal texture, the initial stage of growth is correlated with the formation of dislocation loops characterized by a Burgers vector of $\frac{1}{3}11\bar{2}0$ and a loop plane normal distributed between the first and second order prismatic planes [10–12]. They are either of vacancy or interstitial character, and are commonly referred to as *a* loops. During service strongly textured and recrystallised Zr alloys show some initial

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rapid growth followed by a slow steady state growth phase. The initial growth phase is associated with an increase in total dislocation line density of a loops [13,14], while transmission electron microscopy (TEM) investigations have reported a loop number density saturation during the steady state growth phase [15]. After the period of limited growth a threshold fluence is reached where a second stage of accelerated or "breakaway growth" occurs [6,16]. Breakaway growth in neutron-irradiated Zr alloys has been correlated with the irradiation-induced formation of large faulted vacancy dislocation loops with a Burgers vector of $\frac{1}{6}20\overline{2}3$ that lie on the basal plane [4,17] and are hereafter referred to simply as c loops. The nucleation mechanism for c loops is not fully understood, but is believed to be related to the presence of diffusing solute elements from dissolving precipitates [18].

The characterisation and quantification of dislocation loops in irradiated nuclear reactor materials has traditionally been limited to TEM analysis, as it enables direct imaging of the line defects. However, it is very time consuming to perform statistically reliable measurements of dislocation densities in a TEM, particularly for irradiated materials where it can be challenging to distinguish the dislocation loops from other sources of contrast [11,17]. Therefore, quantitative assessments of dislocation densities from TEM measurements are lacking, especially at high fluences where the a loop density is large and the newly formed c loops tend to be of greater interest. For mechanically deformed materials the development and implementation of diffraction peak profile analysis (DPPA) has facilitated the study of bulk dislocation structure in [19–21]. DPPA allows for indirect, but statistically reliable, measurements of dislocation density and population distribution [22], providing a complementary technique to TEM analysis [23].

In the present study the aim was to investigate the dislocation structure of a set of Zircaloy-2 samples extracted from fuel cladding and channel boxes, irradiated in boiling water reactors to fluences between 8.7×10^{25} n m^{-2} and 14.7×10^{25} n m^{-2} (E > 1 MeV) [24,25], via DPPA, to elucidate the high fluence dislocation evolution and compare the data with conventional dislocation analysis by bright field TEM imaging. In order to achieve this, diffraction profiles were recorded on the I11 beamline at the Diamond Light Source [26], UK, which have been analysed using the extended Convolutional Multiple Whole Profile (CMWP) analysis procedure [19,27–29].

2. Experimental methods

2.1. Material

Westinghouse Electric Company and Studsvik Nuclear AB supplied neutron-irradiated, recrystallised Zircaloy-2 samples [24] in the form of six electro-polished TEM foils. The nominal chemical composition of the material, together with the details of the foils, is presented in Table 1, in order of increasing neutron fluence. Of the six foils, four were extracted from fuel cladding of the Westinghouse designated LK3™ type [30] from the Leibstadt (KKL) boiling water reactor (BWR), one foil was taken from a channel box sample from KKL and one foil was extracted from a channel box sample from the Olkiluoto-2 (OL-2) BWR. The channel boxes are manufactured by following a processing path similar to the cladding, i.e. melting, hot-rolling and β-quenching, followed by a series of cold-rolling/annealing steps culminating in a final recrystallisation anneal.

It is difficult to acquire a good selection of material that has been neutron irradiated in controlled, identical conditions to different fluences. The investigation of cladding and channel together is not ideal, as the two structures are in different neutron environments and experience a temperature difference [31] of approximately 50 °C, however the material presently studied is what was most readily available.

In addition, Westinghouse Electric Company also supplied recrystallised non-irradiated LK3™ type Zircaloy-2 sheet, which was used to prepare a non-irradiated reference bulk sample by standard mechanical grinding and polishing.

The total in-reactor elongation strain data associated with the cladding material is shown in Fig. 1. It is evident from the linear elongation strain of the cladding and the high neutron fluences, all of which are above the 3-4x10^{25} n m^{-2} generally accepted threshold fluence range for initiation of breakaway growth [32], that these samples have experienced breakaway growth. However, it should be noted that the samples came from a functioning fuel assembly and as such was not stress-free during service, therefore the strain represents both irradiation-induced growth and irradiation-enhanced creep as well as strains derived from oxide and hydride formation and pellet-cladding interaction [3,33].

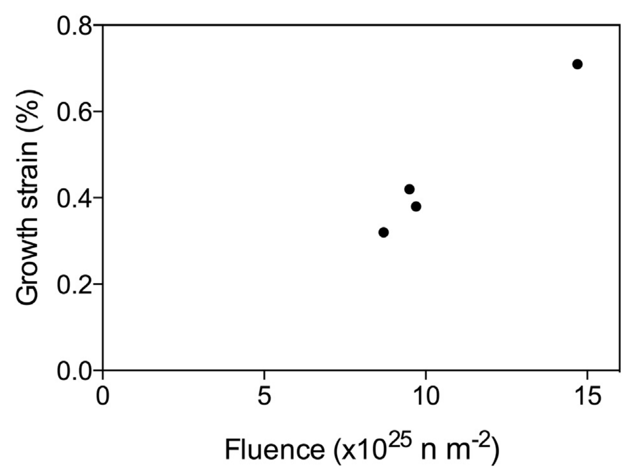

Fig. 1. Growth strain of 4-meter Zircaloy-2 fuel rods derived from rod growth data in Table 1.

Table 1
Neutron irradiated Zircaloy-2 sample data with * indicating cladding samples, ** indicating channel samples from Leibstadt BWR and *** indicating channel samples from Olkiluoto-2 BWR.

Chemical composition (wt.%): 1.34–1.35 Sn, 0.17–0.18 Fe, 0.11 Cr, 0.05–0.07 Ni					
Sample	♯ cycles	Rod average neutron fluence (10^{25} n m-2)	Rod growth (mm)	Approx. core elevation (mm)	Rod average burnup (MWd/kg)
1*	5	8.7	12.7	2700	51.1
2*	6	9.5	16.6	2700	57.3
3*	7	9.7	15.3	1100	57.4
4***	5	11	N/A	N/A	N/A
5**	7	13.1	N/A	N/A	N/A
6*	9	14.7	28.4	2100	78.7